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**Size limited mesoscopic laser plasma accelerators**

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TATA INSTITUTE OF FUNDAMENTAL RESEARCH**

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Final Report**

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# A Novel source of mesoscopic particles for laser plasma studies

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Intense ( $\geq 10^{18}$  W/cm<sup>2</sup>) ultrashort ( $\leq 100$  fs) laser produced plasmas generate and sustain extremely large electric fields ( $\geq 100$  GV/m) that are known for highly compact ion/electron acceleration schemes. Innovative target engineering, particularly for ion acceleration, assumes importance from the perspective of developing university-scale table top laser-plasma accelerators. Nanometric-scale atomic clusters or nanoparticles coated on a solid substrate as targets have been reported to generate multi-MeV ion beams even at moderate intensities accessible in most laboratories. In this context, the study of targets comprising mesoscopic particles of sizes on the order of magnitude of the laser wavelength for laser plasma acceleration is a sparsely explored domain, theoretically and experimentally. Mie-scattering and/or surface plasmon resonances are expected to induce local enhancement of the laser electric field, increasing the efficiency of hot electron generation, a precursor to particle acceleration. In this following report we outline the development of a delivery mechanism of microparticles into an effusive jet in vacuum for laser plasma studies. We characterise the device in terms of particle density, particle size distribution, and duration of operation under conditions suitable for laser plasma studies. We also present the first results of the diagnostics of the laser plasma under relatively mild intensities of  $10^{15}$  W/cm<sup>2</sup>, wherein highest hot electron temperatures of about 370 keV are observed.

## I. INTRODUCTION

Pulsed laser interaction with matter at intensities above  $10^{14}$  W/cm<sup>2</sup> inevitably includes ionization leading to the formation of plasma on timescales of a few hundreds of femtoseconds, often times even before the peak of the pulse has arrived. In short pulse lasers of  $\leq 100$  fs duration, ion motion is negligible, implying that the laser directly transfers energy to the electrons in the plasma. Through a number of mechanisms<sup>1-3</sup> which essentially involve the laser driving the electrons through a strong plasma density gradient, the electrons are heated to temperatures which can even be in the MeV range. The transfer of this energy to the ions takes many forms depending on the nature of the target, which ultimately yields fast ions from the plasma. Over the last decade laser plasma acceleration has made rapid strides in terms of providing high brightness,<sup>4,5</sup> tunable, monochromatic,<sup>6</sup> energetic beams of electrons and ions. The promise of table top systems with applications in cancer therapy, isotope preparation, radiography and thermonuclear fusion, to name a few, has catapulted research in the field of high density, high field science to the forefront in many centers worldwide.

Realizing intensities of the order of  $10^{19}$  W/cm<sup>2</sup> and above is still at the frontier of laser engineering at the moment with only a few centers worldwide being able to provide it. To achieve laser driven ion acceleration on the MeV level, at intensities which are at least 100 times lower would require a different approach. Target design has been an approach showing much promise in

recent times. Hard X-rays, which are an indication of the electron temperature, have been shown to have a near 50-fold enhancement from plasmas created on surfaces with nanometric modifications<sup>7,8</sup> as compared to those on polished surfaces. As revealed by particle-in-cell plasma simulations<sup>8</sup>, depending on the aspect ratio, length and density of the nanostructures on the surface, the enhancement in the intensities can be up to 100 fold. Recent experiments with nanowires<sup>9</sup> report protons of energy 5.5 MeV with moderate intensities of  $5 \times 10^{17}$  W/cm<sup>2</sup>.

Mesoscopic particle targets evoke further interest in this regard as structures comparable to the wavelength of the laser can lead to 10-100 times enhancement in the fields through Mie resonances. Such enhancements have already been shown in plasmas created in micron-scale liquid droplets<sup>10,11</sup>. Studies with mesoscopic particles have been limited to microdroplets, or surface modifications by arraying microspheres on substrates<sup>12,13</sup>. The limitation with microdroplets is their spherical shape, which cannot be modified any further. Mesoscale targets with nanometric modifications, either in terms of having spheroid shapes or even pointed ends may see further enhancements due to the so-called lightning-rod effect. Indeed with spheroid shaped *E. coli* bacteria with sizes of the order of  $1 \mu\text{m}$ , a large enhancement of two orders in magnitude in the x-ray yield has been observed<sup>14</sup> at modest intensities ( $\geq 10^{17}$  W/cm<sup>2</sup>). These target are invariably used as a coating on plain solid substrate. Plasma generation in the solid substrate and the electron transfer from the bulk solid, changes the dynamics of the interaction and makes it difficult to clearly decipher the novelty of the laser plasma generation in the mesoscopic particles. Large continuum of free electrons on a scale much larger than the laser wavelength results in reflection of a substantial fraction of light and subsequently less coupling of the incident light to plasma generation.

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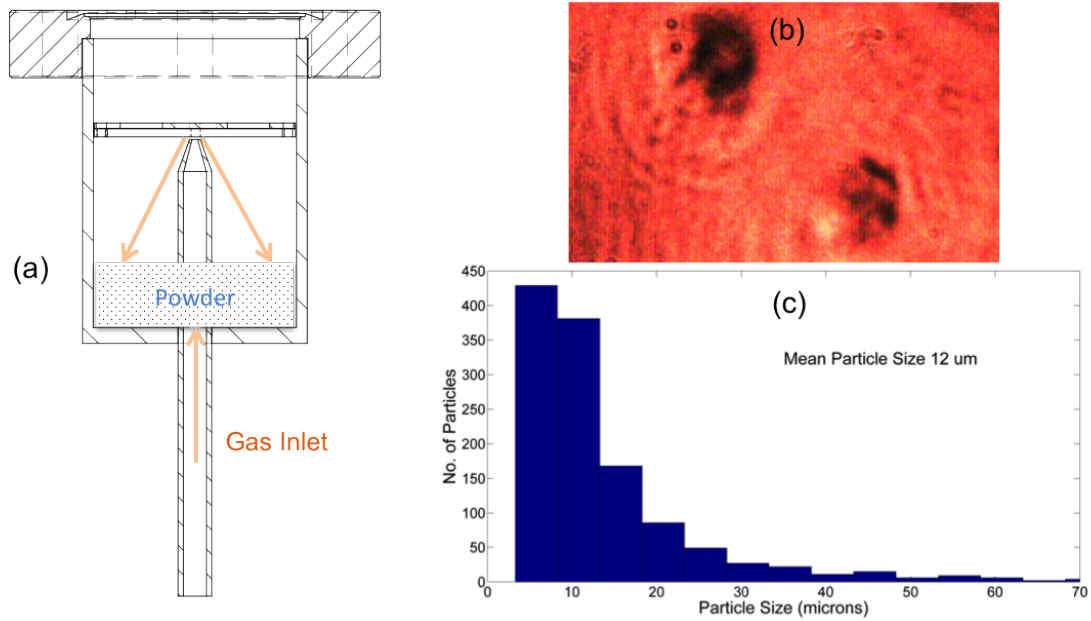


FIG. 1. (a) The design of the first prototype device. (b) A typical shadowgraph showing two particles in the image. (c) The histogram of particle sizes obtained.

The charge densities achieved in laser produced plasma with a solid substrate are much smaller than that with the solid-density sub-wavelength particles. To explore the physics of laser absorption and its optimal use in x-ray generation or particle acceleration, it is therefore necessary to design an experimental apparatus that can deliver meso-scopic particles from a pre-engineered particulate powder. The ability to synthesise particles of any size, shape and material compositions using conventional solid-state synthetic methods can come handy in generating designer targets for efficient plasma generation. Such target systems are crucial not only for understanding the basic intense laser plasma science but also for manipulating the intense laser produced plasmas for efficient x-ray generation or particle acceleration.

In this submission we have considered a novel approach to study laser plasmas on the mesoscopic scales, which is to develop a mechanism to deliver sub-micron to micron size particles as an effusive jet into vacuum. It offers an effective way of replenishing solid targets, particularly important in the context of high-repetition lasers. Additionally, the size limited nature of the particles requires that energy dissipation mechanisms which are self-evident in substrates are not present here. The strategy we employed for generating a beam of microparticles in vacuum akin to particle seeding used in particle image velocimetry<sup>15</sup>. Pulses of pressurised gas passing through a chamber of pre-synthesised particles can form an aerosol mixture of the particles in the gas. The jet of particles carried by the gas then interacts with the focussed laser beam in vacuum. We describe the design of such a particle delivery mechanism and characterise the

particle density, particle size distribution, and duration of operation of the device. We also present the first results from the laser plasma produced by such microscopic particles.

## II. PROTOTYPE DESIGNS

Seeding particles into a gas stream to create aerosol mixtures essentially involves entrainment of particles from a powder into the gas, either by "bubbling" the carrier gas through a fluidized bed of particles or through pickup by 'cyclone'-type turbulences<sup>15</sup>. In Fig. 1(a) is shown a sketch of the first prototype device tested for suspending the particles into the gas jet. Compressed air is introduced into the chamber containing the powder through a 1/4 inch tube. The exit of this tube is constricted to a 1 mm orifice. The flow of the gas is interrupted by a baffle which directs the flow onto the powder. The gas impacting the powder surface lifts particles into the gas, which are carried to the top of the chamber. A short tube at the exit of the chamber carries the aerosol into a vacuum chamber. When the carrier gas is pulsed with the help of a solenoid throttling valve, a pulsed effusive jet of particles is obtained (gas pulsing is essential to maintain the vacuum level).

To visualise the particles, a shadowgraph based imaging set up was commissioned. A pulsed (10 us) LED source (lab-built) was used to illuminate the gas jet. An imaging system with a magnification of 8.5X was used to project the particle images onto a triggered CCD camera. The pulsed gas jet, laser and camera trigger

were synchronised with a common Stanford Research system (SRS) function generator to obtain shadowgraphs of the particles with each pulse. Nitrogen gas was pulsed through a throttling nozzle of 750  $\mu\text{m}$ , using a Series 90 Parker valve, with a stagnant pressure of 10 bars. At a repetition rate of 5 Hz, gas pulse opening times of 5 ms, a sample of the images obtained is shown in Fig. 1(b). The particles seeded were from commercial grade Boric Acid ( $\text{H}_3\text{BO}_3$ ) powder, baked and cooled before operation. The shadowgraph images were processed in MATLAB to extract the number of particles in each image and their sizes. The histogram of particle sizes obtained from a typical run is presented in Fig. 1(c), wherein the particle size is the square root of the area of the particle (a good metric for irregular shaped particles). From the area of the image frame (magnification) and with an average of 2 particles per image frame, we estimate a particle density of  $3 \times 10^4 / \text{cm}^3$ .

While, the first prototype produced promising results, particularly in terms of particle densities in the jet the major limitation of the design was found to be the very short duration of operation of only around 5 minutes. Such short lifetimes of the particle jet is unusable for laser plasma studies, particularly when it required to build statistics in charged particle and photon spectra over a large number of laser shots. To delve into the cause for the limited particle beam lifetime from the machine, a series of observations were conducted, by varying configurations of the gas inlet and baffle positions, and inspecting the residual powder in the chamber at the end. It was observed that invariably, over time, the powder would redistribute within the volume, with channels or depressions, to allow a free unhindered path for the driving air. So even with sufficient powder reserve in the chamber, the particle flux within the air would reduce drastically, as the driving gas would not encounter any particles.

An improved version was conceived to counter this effect. The first change was to move from the hitherto 'cyclone' based entrainment system to the mechanism of passing the driving gas through the bulk of the powder. While this increases the particle flux, it does not ensure that over time gas channels are not formed in the bulk of the powder. Essentially, an alternate system is required to ensure 'fluidity' of the bulk of the powder. We therefore introduced a rotating blade (lower panel of Fig. 2) driven by an external motor (100 rpm) which would prevent gas channels from forming in the bulk of the powder. To characterise the duration of operation of the new version a Mie-Scattering setup was arranged at the particle exit. For particles of the size comparable or larger than wavelength of light scattered, i.e., typically 0.5-1 microns, Mie-Scattering leads to increased scattered photon flux in the direction of the light propagation. In the set up a 650 nm CW laser beam was shined through the particle jet. The particle jet was pulsed by pulsing the driving gas, and the pulses of scattered light was recorded over time by a photodiode at a very small angle

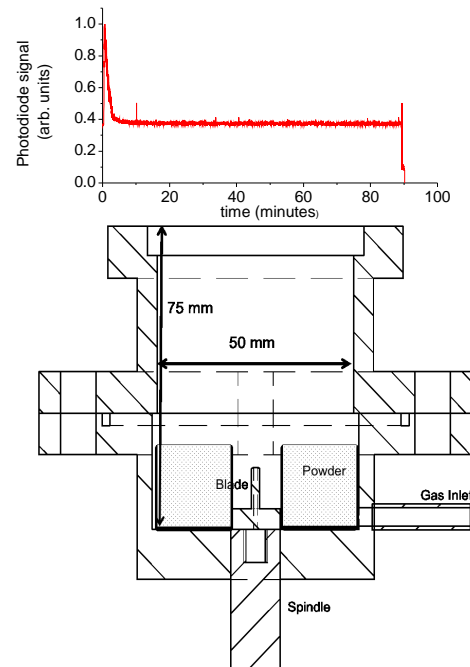


FIG. 2. A sketch of the second prototype device. On the top is a plot of the peak values of the Mie-scattered signal from the particles captured by a photodiode, as a function of time in minutes. The driving gas pulses (see text) are at 8 Hz, 10 ms, with a stagnant pressure of 10 bar, through a 0.75 mm dia. nozzle

$\approx 5$  degrees to the incident beam, along the direction of propagation. In the top panel of Fig. 2, the peak values of the photodiode pulses is plotted a function of time in minutes. A very sharp rise of the signal at the beginning in a time duration of around 2 minutes is followed by a settled signal level which remain constant over an hour. Additionally, the shot to shot stability in the settled period is very good with a standard deviation of 2 percent of the median.

### III. FIRST RESULTS FROM SIZE-LIMITED MESOSCOPIC LASER PLASMAS

With the assurance that stable operation of the delivery mechanism for extended durations is possible, the system was then coupled with a customized vacuum chamber to initiate interactions with intense femtosecond laser pulses. A stainless steel tube at the exit of the chamber guided the aerosol into a cubical vacuum chamber of 125  $\text{cm}^3$ . The other end of the tube ended into a receiving vacuum chamber that acted as a trap for the particles (for recycling the powder and to prevent contamination of the vacuum pump). Four 2 mm holes were drilled into the tube, two corresponding to the entry and exit of the laser and two perpendicular to these for diagnostics of the laser plasma. A rotary pump evacuated

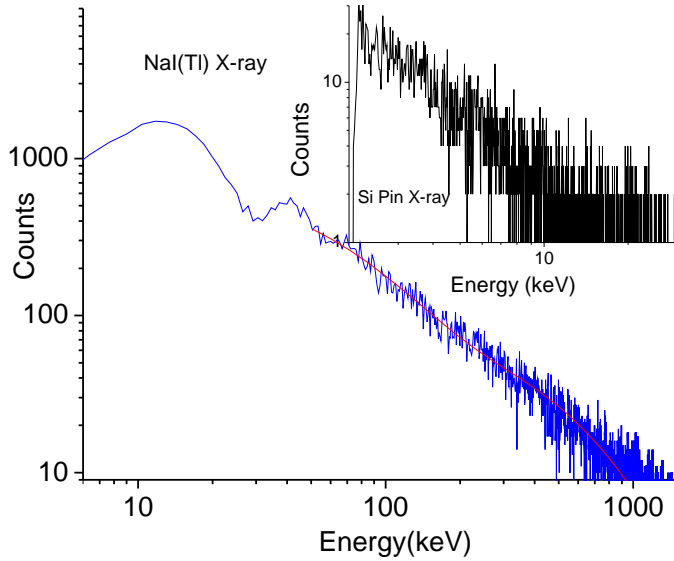


FIG. 3. Hard X-rays from the bremsstrahlung of hot electrons from the laser plasma formed from Boric acid particles in the jet measured by a NaI(Tl) s X-ray detector. The red curve is the two-electron temperature fit, assuming a Maxwellian distribution. The inset shows the low energy X-ray spectra captured by a Si PIN detector.

the vacuum chamber to base pressure conditions of 0.05 Torr. Under typical experiment conditions, with stagnant pressures of 11 bar, throttling valve opening time of 0.9 ms, and 5 Hz repetition rates, the operating pressure rose 0.5 Torr.

30 fs, 2.7 mJ laser pulses were focused in a  $f/5$  geometry into the boric acid particle beam, to a measured  $1/e^2$  beam spot size of  $\approx 70$   $\mu\text{m}$ . This results in a moderate intensity of  $2.9 \times 10^{15}$  W/cm<sup>2</sup>. The measures of the thermal and hot electron temperatures offer conventional diagnostics of the transient laser plasma. The soft and hard X-ray spectrum from bremsstrahlung of the plasma electrons gives a direct measure of this. To this end, X-rays through a Mylar window were captured by a Si pin detector (XR-CR 100, Amptek Inc.) and recorded by an MCA (model 8000, Amptek Inc.). The angle intercepted by the detector was around 20 degrees, at a distance of 5 cm. The hard X-ray in the range of 30-1000 keV was also collected by a NaI (Tl) detector placed about 5 cm from the interaction region. The detector was placed at increasing distances, reducing the solid angle of collection. At all distances (varied count rates) the spectral characteristics were similar, indicating the absence of pile-up effects in the detector. Further background cosmic ray spectra was also acquired for the same amount of time as the x-rays from the laser plasma, which is about 30 minutes. The background subtracted data is presented in Fig. 3, the inset showing a typical spectrum from the Si PIN detector. Assuming a Maxwellian distribution of the

electrons in the plasma, we fit the bremsstrahlung temperature for the hard X-rays over 50 keV (for which we can assume 100 percent transmission through the glass and metal). A two temperature fit is evident, with temperatures of  $51.6 \pm 0.7$  keV and  $370 \pm 6$  keV (about 20% weightage).

At comparable intensities with and additional 10 ns pre-pulse, the hot electrons from 15  $\mu\text{m}$  micro droplets have energies of  $\approx 36$  keV<sup>11</sup>, but do not show evidence of a high energy tail. The results from the micro particle jet are impressive when one compares them to those obtained from micro metric<sup>14</sup> and nanometric<sup>8</sup> modifications on substrate, wherein the electron temperatures recorded are of the order of 50-70 keV, albeit the intensities being an order of magnitude higher. Furthermore, the evidence of an unusually high energy tail in as seen in the aerosolised particles have not been observed hitherto, particularly at such moderate intensities. These preliminary results already raise interesting questions on the mechanisms of energy transfer to mesoscopic laser plasmas. Since both geometry and size are expected to play a major role in the coupling dynamics, the new delivery mechanism offers a unique opportunity to study these effects in detail by systematically varying the size and geometry of particles in the aerosol.

#### IV. OUTLOOK AND FUTURE STUDIES

The mechanism for delivery of mesoscopic particles for laser plasma studies has shown potential for opening new lines of enquiry into this unexplored area. The studies presented here are a proof of principle of the mechanism and its utility to laser plasma experiments. Current work is focussed on adding further diagnostic capability such as optical spectroscopy, ion and electron spectroscopy. With these in place, systematic variation of size and shape, with pre-synthesised mono-disperse particles can be studied under great detail. Furthermore, adaptations of the device to conform to the target chamber at Tata Institute of Fundamental Research, Mumbai is also underway to enable experiments at and near relativistic intensities ( $10^{18}$ - $10^{19}$  W/cm<sup>2</sup>).

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## **Executive Summary**

**PI Name:** M.Krishnamurthy

**Title of Project:** Size limited mesoscopic laser plasma accelerator

**What significant findings came from this project:** We have designed, developed and characterized a effusive jet source that entrails particles pre-engineered and made in the form of a powder. The particles can be nano-metric or micrometric in size. In this source individual particles are suspend in vacuum and can be subjected to ultrashort light at intensities as large as  $10^{19} \text{ Wcm}^{-2}$ . The source has large enough particle density that there is at least one particle in the focal volume for each laser pulse of a laser (tested with a laser that operates at 1 kHz). Imaging in vacuum clearly shows that the methodology delivers individual particle with out any agglomeration. This provides a novel target source for studying high energy density science with particles of any size, shape and material composition. This is crucial for the basic understanding of the absorption mechanisms of how intense laser light is absorbed by particles of solid-like density but size that is nano-metric (much smaller that laser wavelength) or micrometric (about the laser wavelength or a few times higher).

**What is/are the significance of the findings:** The first experiments on the high energy density science with  $20 \mu\text{m}$  boric acid particles exposed to  $10^{15} \text{ Wcm}^{-2}$  intense light (formed by focusing 2.7mJ, 30 fs pulses) show an unprecedented x-ray emission with an electron temperature of about 50 keV and higher energy component of about 370 keV. The x-ray energies extend up to an MeV and achieving this with modest laser energies is significant in shaping the intense laser produced plasma as efficient compact x-ray source and/or electron accelerator.

**What new research questions came about from this project:** The findings mentioned above are, to the best of our knowledge, the highest electron temperatures achieved in intense laser produced plasmas at such small intensity. The present lasers operate at 1kHz repletion rate but commercial system at ten times higher repetition rate laser are available. X-ray source of such high energy and such a high repetition rate is a very significant finding in the intense laser plasma science. We need to expand on these first results and do many measurements with this source that gives the option to vary the size, shape and material compositions of the particles that absorb the laser light. Understanding the underlying mechanism will be a crucial advance in the x-ray generation science with intense laser plasmas.

## Final Report for AOARD Grant 144023 “Size limited mesoscopic laser plasma accelerator”

1 Nov 2015

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**Abstract:** Intense laser produced plasma are known for generating high dense - high temperatures plasma that is a source for electron, ion acceleration and also x-ray emission that extends to a few MeV in energy. Size limited particles in the nm and micro meter range are known to absorb the laser light efficiently. The present project has fulfilled the aim of developing a source of nearly monodisperse microparticles delivered as an effusive jet into vacuum for laser plasma studies. This instrument extends the range and scope of the nature of particles that absorb the laser energy and provide a richer variety of micro-nanoplasma environment for not only basic studies of understanding the intense laser field science but also to provide new technologies for developing efficient x-ray sources. After due characterization of the source, the instrument was used for studying x-ray generation ( $<10^{16}$  Wcm<sup>-2</sup> intensity range). The first results show dramatic effects in x-ray generation from the size limited particles. X-ray emission measurements from focusing 2.7mJ-30fs laser pulses on boric acid particles on about 15  $\mu$ m size, demonstrate an electron temperature of about 50 keV with a high energy component (of 20% weightage) of about 370 keV. This is perhaps the highest electron temperature achieved in laser produced plasmas formed with a few mJ-fs pulses. A detailed investigation of the underlying physics especially on the role of the size and shape of the target is currently under exploration.

**The detail elements of the work completed that would constitute the report are included in the draft of a paper that is presently being prepared for submission to the Rev. Sci. Instrum. Journal for a peer reviewed publication.**

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